Improved Measurement of the $1s2s\,^1S_0 - 1s2p\,^3P_1$ Interval in Heliumlike Silicon

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Using colinear fast-beam laser spectroscopy with copropagating and counter-propagating beams we have measured the $1s2s\,^1S_0 - 1s2p\,^3P_1$ intercombination interval in $^{28}Si^{12+}$ with the result 7230.585(6) cm^{-1}. The experiment made use of a dual-wavelength, high-finesse, power build-up cavity excited by single-frequency lasers at 1319 and 1450 nm. The result will provide a precision test of *ab initio* relativistic many-body atomic theory at moderate Z.

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As the prototypical multielectron atomic system, helium and heliumlike ions have long provided a testing ground for the development of *ab initio* atomic theory [1–11]. While nowadays the nonrelativistic Schrödinger equation for helium can be solved to such high accuracy that the results can be considered exact for all practical purposes [4], the *relativistic* two-electron atom is far from a fully-solved problem. The underlying physics, quantum-electrodynamics (QED), is well known, but how to formulate the problem to obtain accurate numerical predictions remains the subject of extensive theoretical effort. For moderate Z ($Z\alpha \sim 0.1$) the problem is the most general and the most challenging since electron-electron correlation must be treated to high order and yet QED effects are also large. Electron correlation effects derived from a Hamiltonian can now be treated accurately using relativistic many-body perturbation theory (MBPT) and equivalent methods [5–8]; however, QED predictions must be derived from fully covariant techniques [5,9,10], and the combination is difficult to treat in a systematic way. A promising method for merging MBPT and QED, called the *covariant evolution operator* method, has been discussed in Ref. [11]. Immediate motivation for an improved synthesis of methods that accurately treat many-body effects and bound-state QED is provided by the current discrepancies between different theory, and between theory and experiment, for the $1s2p\,^3P$ fine structure intervals of He—the theoretical uncertainty frustrates a major effort to obtain an atomic-structure-based value for the fine structure constant for comparison with that obtained from the g factor of the free electron [12–18], and also by proposed experiments at very high Z, aimed at testing QED in strong Coulomb fields, e.g., see Ref. [19].

As reviewed recently in Ref. [10], spectroscopic measurements with sufficient precision to challenge even the existing theory are sparse for heliumlike ions with $Z\alpha \sim 0.1$ [20,21]. Compared to other transitions in moderate-Z heliumlike ions, the $1s2s\,^1S_0 - 1s2p\,^3P_1$ interval has the important experimental advantages of lying within the laser-accessible infrared, of being partially electric-dipole allowed, and of being very sensitive to theoretically interesting QED contributions [22,23]. In Ref. [24] we reported a measurement of this interval in $^{28}Si^{12+}$ with sufficient precision to differentiate between various theoretical values. Since improved theoretical results are now anticipated we have carried out a remeasurement, improving the precision by a factor of more than 30, which we report here. Our new result has an uncertainty equivalent to $2.5 \times 10^{-4}(Z\alpha)^4$ atomic units (a.u.), 13 parts-per-million (ppm) of the total QED correction, or 0.25% of the nuclear size correction [5] and provides a test of theory for heliumlike ions with unprecedented sensitivity.

The experimental arrangement derives from that described in Refs. [24,25]. Here we provide a brief summary before describing the changes leading to improved precision. Beams of $^{28}Si^{5+}$ ions, approximate energy 29.1 MeV ($v/c = 4.7\%$), were obtained from the Florida State University Tandem Van de Graaff accelerator, fitted with a recirculating terminal gas stripper. After momentum analysis in a 90° bending magnet, the ion beam was focused into the interaction chamber where it was collimated to a cross-section of 1 mm × 1 mm, with a typical current of 10 particle nA, and then directed through a nominal 4 $\mu g/cm^2$ carbon foil. The emerging beam had a Si^{12+} fraction of around 7%, of which ~1% was in the $1s2s\,^1S_0$ state, with mean lifetime of 11.5 ns [26]. The Si^{12+} charge state was then magnetically deflected by approximately 5° to be colinear with the optical axis of the high-finesse power build-up cavity (BC). At a distance 25 cm down-beam of the foil, the ions passed through the waist of the laser-excited TEM_{00} mode of the BC, which was 1.9 cm above the 2 cm × 3 cm window of a proportional counter, optimized to detect x-rays from the decay of $1s2p\,^3P_1$ to the ground state at 1.85 keV. The signal was the increase in x-ray count rate due to laser-induced transitions from $1s2s\,^1S_0$ to $1s2p\,^3P_1$, followed by the decay to the ground state of the $1s2p\,^3P_1$ level, mean lifetime 6.36 ps [27]. The resonances were scanned at fixed laser frequency by varying the beam velocity, and hence Doppler-shift, by stepping the field in the 90° magnet, which was monitored by an NMR probe. The accelerator terminal potential was stabilized to follow this magnetic field. To reduce the rate of foil damage and thickening, the foil was continuously scanned vertically through the ion beam, and the vacuum was maintained at $\sim 1 \times 10^{-6}$ mbar.

The BC consisted of two ultra-high-reflectance mirrors, 1 m radius of curvature, spaced 92 cm apart using 4 invar rods inside the vacuum chamber. The mirrors were coated for high reflectance at both 1319 and 1450 nm with measured transmissions at these wavelengths of 55 and 35 ppm. To excite the BC at 1319 nm we used a 300 mW continuous-wave monolithic Nd:YAG laser as before, but for 1450 nm we developed our own single-frequency laser system. This was based on fiber-coupled diode lasers designed as pump lasers for amplifiers used in telecommunications (FITEL inc). The first stage of the 1450 nm laser system consisted of one of these diode lasers spliced to a fiber-Bragg-grating (FBG), forming a 16.5 cm long, in-fiber extended cavity, which enabled single-mode lasing. Although this laser has been operated with a single-mode output of 140 mW [28], we achieved greater stability at the wavelength required for our experiment with a reduced injection current and an output of 30 mW. This was used to injection lock, via a free-space opto-isolator, a second fiber-coupled diode laser. The second laser, which was conservatively operated at 800 mA, yielded an injection-locked output of over 200 mW, of which nearly 100 mW was input into the BC. The FBG-stabilized master laser could be tuned over a range of 500 MHz by varying the voltage applied to a piezo that strained the fiber between the FBG and diode laser, and also by changing the temperature of the FBG, with a tuning coefficient of -1.8 GHz/K. The lock to the ~ 3 kHz FWHM TEM_{00} modes of the BC used the Pound-Drever-Hall (PDH) technique with feedback both to the piezo and the injection current of the master laser [29].

During the data taking the estimated maximum circulating power in the BC was 770 W at 1319 nm and 650 W at 1450 nm. For both wavelengths the BC power was modulated at 500 Hz, with modulation depth >80%, by overdriving the electro-optic modulators used in the PDH scheme. Hence we obtained the laser-induced increase in x-ray count rate, which we normalized to the x-ray background and BC power, as a function of magnetic field in the analyzing magnet. This magnetic field, to a good approximation, is linearly related to the relativistic momentum of the Si^{5+} beam, and hence, except for an estimated 90 keV energy loss in the foil, to the relativistic momentum of the Si^{12+} beam. The laser frequency was continuously monitored using a scanning Michelson wave meter (Burleigh Instruments). Immediately after the Si^{12+} data taking, the wave meter was calibrated by admitting <1 mbar of water vapor into the interaction chamber and using the BC for cavity-enhanced absorption spectroscopy of weak $H_2{}^{16}O$ absorption lines. The calibration lines used have been accurately measured using a Fourier-transform infra-red spectrometer by Toth [30], and have quoted wave numbers of 6895.651, 6896.5230, 7579.5075, and 7579.7831 cm^{-1}, all with errors <0.001 cm^{-1}.

The improvement in precision compared to Ref. [24] was obtained by inducing the $1s2s\,{}^1S_0 - 1s2p\,{}^3P_1$ transition using the wave in the BC counterpropagating with respect to the ion beam, with the BC excited by the 1450 nm laser, alternately with the copropagating wave with the BC excited by the 1319 nm laser. Because of the opposite Doppler shifts, this resulted in resonances at nearly the same beam velocities with similar line shape. Using the relativistic Doppler formula, the transition frequency in the ion frame, ν_0, is related to the laboratory frequencies of the copropagating and counterpropagating lasers, ν_1, ν_2, to more than adequate precision in the present case [31], by ${\nu_0}^2 = \nu_1\nu_2\{1 + \Delta p(1 - p^2/2) + \theta^2 p^2 - \Delta(\theta^2)p/2\ldots\}$, where $\Delta p = p_2 - p_1$, $p = (p_1 + p_2)/2$, $\Delta(\theta^2) = {\theta_2}^2 - {\theta_1}^2$, $\theta^2 = ({\theta_2}^2 + {\theta_1}^2)/2$, where $p_{1,2}$ and $\theta_{1,2}$ are the values of $\beta\gamma$ and the (small) angles of intersection of the ion and laser beams, corresponding to the centroids of the respective resonances. Hence only the small difference in $\beta\gamma$, Δp, proportional to the change in the magnetic field in the analyzing magnet, ΔB, must be accurately measured. By contrast, in Ref. [24], where resonances were induced with a copropagating 1319 nm laser only, it was necessary to determine the absolute beam velocity by calibrating the analyzing magnet and estimating the energy loss of the Si^{12+} ions in the foil.

The resonance data for our final result were obtained over three days. Each day, after careful alignment of the ion beam to the optical axis of the BC, the run consisted of scans of the analyzing magnet, and hence ion beam velocity, in both directions across the $1s2s\,{}^1S_0 - 1s2p\,{}^3P_1$ transition, with the cavity excited first with the 1450 nm laser, then the 1319 nm laser, and finally with the 1450 nm laser again. By averaging centroids of the 1450 nm induced resonances on each day we took account of possible shifts due to foil thickening. On the first and third days the lasers were set to frequencies such that the copropagating and counter-propagating resonances occurred at nearly the same value of the analyzing magnetic field, approximately 9460 G. On the second day, the laser frequencies were offset so the resonances were separated by $\Delta B \approx 44$ gauss. The laser-induced x-ray resonances are similar to those shown in Fig. 2 of Ref. [24], but with reduced signal-to-background, typically 2% at the peak, due to the lower power in the BC. This was partially compensated for by taking more data near the half-height points. These resonances were fitted with convolutions of a Lorentzian, representing the natural width of 25 GHz, and a narrower, phenomenological "Gaussian-with-exponential tail", to represent the Doppler profile due to the velocity spread of the Si^{12+} ions leaving the foil. It is important to note, that since only the difference in magnetic field, ΔB, between the 1319 and 1450 nm induced resonances must be accurately measured, the small asymmetry in the Doppler-tuned line shape, since it is common to both resonances, does not affect our result to first order.

Our analysis proceeds by using the fits to the resonance data to obtain the best values of ΔB between the 1450 and 1319 nm resonances for each day. Then, assuming $\Delta(\theta^2) = 0$ and $\theta^2 = 0$, and values for p and the differential magnet calibration $d(\Delta p)/d(\Delta B)$ from the work of Ref. [24], we obtained the three values for the transition frequency ν_0, which are shown in Fig. 1.

Based on our previous experience with this analyzing magnet it is likely that the fractional uncertainty in the differential magnet calibration used to obtain the values for ν_0 in Fig. 1 is <0.2%, in which case we could simply average the three results. However, instead, we take account of any error in our assumed magnet calibration by simply extrapolating the three points to $\Delta p = 0$. The result, converted to wave number, is $\nu_0 = 7230.5852\ \mathrm{cm}^{-1}$ with statistical uncertainty $0.0054\ \mathrm{cm}^{-1}$.

The divergence of the ion beam and its alignment with respect to the axis of the BC were determined by scanning the ion beam across a set of slits, while monitoring the transmitted beam current, and across the mode of the BC while monitoring the laser-induced signal. From these scans, and the measured deflections produced by the steering magnets on our beam line, we determined, for the ion beam averaged across the laser mode, $\langle\theta^2\rangle = (4.2\ \mathrm{mrad})^2 \pm (3.2\ \mathrm{mrad})^2$, and $\langle\Delta(\theta^2)\rangle = (\Delta p/p) \times (26\ \mathrm{mrad})^2 \pm (26\ \mathrm{mrad})^2$. From this it follows that the Doppler-shift errors to ν_0 from misalignment and divergence are below 0.1 ppm and are negligible. A number of other possible sources of systematic error in our Doppler-tuned method were also considered and found to be negligible. These include the correction to the Doppler-shift due to the cavity-induced modification of the optical phase [23] and also the bias to the resonance line shape due to the relativistic variation of the laser intensity seen by the moving ions [32]. Consideration was also given to bias due to variation with beam velocity of the yield of the Si^{12+} ions in the $1s2s\,^1S_0$ state, and of those states responsible for the x-ray backgrounds, and also bias due to transverse walk-off between the ion and laser beams. Except for the relativistic variation in laser power, which in any case produces a negligible shift, all these effects bias the centroid $\beta\gamma$ values of the copropagating and counterpropagating resonances similarly, and the resulting effect on ν_0 is negligible. Shifts due to the ac-Stark effect, and also due to the Zeeman and motional Stark effects due to stray magnetic fields near the detection region are also negligible. Biases due to velocity-dependent depletion of the $1s2s\,^1S_0$ population as the ion and laser beams merge were also estimated to be negligible. Table I summarizes the main sources of error in our result. Besides statistics, the main contribution is from the uncertainty in the calibration and reproducibility of our wave meter.

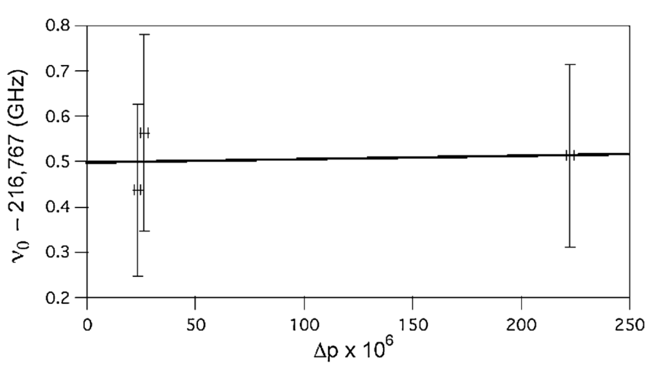

FIG. 1. Results for the $1s2s\,^1S_0 - 1s2p\,^3P_1$ interval in $^{28}\mathrm{Si}^{12+}$, ν_0, versus the difference in $\beta\gamma$, Δp, between the resonances induced with the two lasers. The errors on the points are statistical errors derived from the fits to the resonance curves.

Our final result for $1s2s\,^1S_0 - 1s2p\,^3P_1$ transition in $^{28}\mathrm{Si}^{12+}$ is hence $7230.585(6)\ \mathrm{cm}^{-1}$. This is compared with the previous experiment and theory in Table II.

Our new result is in good agreement with the previous measurement [24] and is considerably more precise than any of the existing theoretical results. Of these, the closest is the result of combining an "all-orders" relativistic MBPT calculation by Plante *et al.* [8], and a QED calculation by Drake [5]. We note, for high Z, ref. [10] represents a significant advance over Refs. [5–8], by providing for the first time QED corrections complete to order $(Z\alpha)^4$ atomic units. Unfortunately, at $Z = 14$, due to insufficiently accurate treatment of correlation, the results are no more precise than the earlier work. Improved calculations of both correlation and QED effects are anticipated [33].

The precision of our measurement is mainly limited by the signal-to-background ratio and the width of the $1s2p\,^3P_1$ level and could be further improved. However, our present uncertainty already approaches the contribution to the theoretical uncertainty from uncertainty in nuclear charge distribution and polarizability [34], which may be considered the limit of the pure atomic physics problem. We also note, because of the rapid increase with Z of the x-ray decay rates of the $1s2p\,^3P_1$ and $1s2s\,^1S_0$ states, and of the background-producing $1s2p\,^3P_2$ state [23], it

TABLE I. Contributions to the final measurement uncertainty.

Source	Uncertainty (ppm)
Statistics	0.75
Line shape asymmetry	<0.1
Ion beam divergence and misalignment	0.02
Yield dependence on velocity	<0.03
Wave meter calibration and reproducibility	0.24
Total	0.79

TABLE II. Experiment and theory for the $1s2s\,^1S_0 - 1s2p\,^3P_1$ transition in $^{28}Si^{12+}$.

Source	Wave number (cm^{-1})
This experiment	7230.585(6)
Redshaw and Myers, experiment [24]	7230.5(2)
Plante *et al.* [8], and Drake [5]	7231.1
Cheng *et al.* [6,7] and Drake [5]	7228.9
Artemyev *et al.* [10]	7229(2)

will be difficult to extend precision laser spectroscopy of this transition to significantly higher Z.

In conclusion, we have measured the $1s2s\,^1S_0 - 1s2p\,^3P_1$ interval in $^{28}Si^{12+}$ using a fast-beam laser technique, with copropagating and counter-propagating beams to cancel the Doppler-shift, obtaining a 30-fold improvement in precision. Our result provides a precision datum for testing developments in the theory of moderate-Z two-electron ions, including efforts to systematically combine MBPT and QED.

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